First Principles Calculations of the 3-k Magnetic Structure and 3-k Distortion in UO_2

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The occurrence of multi-k magnetic structures is often observed in the actinide compounds. These structures may be found in highly symmetric lattices, such as the face-centered cubic (fcc) lattice, in which the star of the wave vector k of the magnetic structure contains several members. If three members of the star are needed to describe the magnetic structure, the magnetic ordering is called 3k. Such is the case in UO_2, which crystallizes in the fcc CaF_2 type structure (see Fig. 1) with a lattice constant of 5.47 Å. UO_2 is a semiconductor with a gap of 2 eV [1,2].

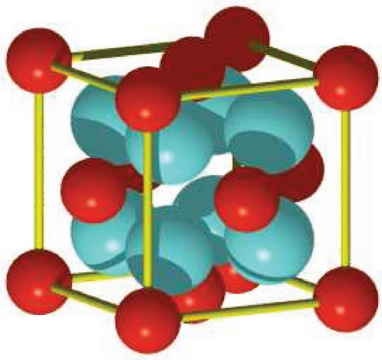


Fig. 1.
The fluorite crystal structure of UO_2. The uranium ions are represented by the red balls. The oxygen ions are placed at a(1/4,1/4,1/4) and a(3/4,3/4,3/4) respectively and are depicted as blue balls.

A first order phase transition [3] takes place at 30.8 K from the paramagnetic state towards a transverse type-I antiferromagnetic (AFM) structure with a magnetic moment of 1.74 μ_B[4]. Crystal field calculations [5] in UO_2 predicted that the ground state of the U^{4+} ion is the Γ_5 triplet. Later, Allen [6, 7] developed a microscopic theory of the effect of spin-lattice interaction on the ground state and spin wave excitations in UO_2. In this theory, the competition between the magnetic superexchange and an effective quadrupolar interaction caused by the spin-lattice interaction results in a non-fully polarized spin of the ground state. In particular, this theory successfully explains the order of the phase transition as well as the reduction of the saturation ordered moment from $2\mu_B$, characteristic of the Γ_5 triplet in the absence of quadrupolar interaction, to the observed value.

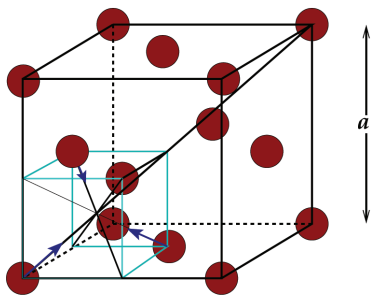
In order to investigate the oxygen distortion [6] that Allen had predicted to turn up at the phase transition, Faber and Lander [4, 8] performed a neutron diffraction study. They deduced from their data that below the Néel temperature the oxygen ions in UO_2 are indeed shifted from their ideal fluorite positions. However, the observed distortion was not the one suggested by Allen. Similarly, their results indicated that the magnetic structure was the 2k structure contrary to the previously reported AFM (1k structure) [3].

Though the agreement was not perfect, neutron scattering experiments under an external magnetic field [9] found that the 3k magnetic structure fitted their data better than the 2k and the 1k magnetic structures. Recently, an inelastic neutron scattering with polarization analysis [10] explored the evolution of the magnetic response through the phase transition. A scenario arises in which uncorrelated 1k dynamical Jahn-Teller distortions occur above the Néel temperature T_N along the three directions of the <100> star and as T_N is approached a correlation develops between the phases of the corresponding vibrations until, eventually a static 3k distortion is obtained at T_N. First principles calculations [11] investigated different multi-k magnetic structures with their corresponding type of oxygen distortion using LDA+U. They determined that the 1k structure is almost always favored, except when they employed the AMF-DDC ("around mean filed" double counting corrections) implementation

of LDA+U, for values of U greater than 0.35 Ry. Nevertheless, their electric field gradients calculated by assuming a 3k model agreed with experiments, with a somehow larger oxygen displacement.

Our present study is motivated by the partial success of theory to determine the magnetic structure and the associated oxygen distortion in UO_2. We have performed first principles calculations in UO_2 with the full-potential augmented plane-wave with local orbitals [12, 13] (FP-APW+lo) method. One of the challenges in describing correctly the ground state of the actinide compounds is the way the 5f electrons are treated. There is a general consensus that the 5f states are well localized in UO_2. Therefore, we used in our calculations the local spin density approximation (LSDA) as parameterized by von Barth and Hedin [14] together with the self-interaction correction (SIC) [15]. Our method is specially suited to study noncollinear structures, such as the 3k observed in UO_2, since the magnetization density is treated as a vector field, i.e., there is no constraint in the shape nor in the direction of the magnetization. We used in our calculations the experimental lattice constant.

We have investigated the 3k magnetic structure (see Fig. 2) together with the



3k oxygen distortion proposed by Burlet et al. [9] and compared with the simple 1k magnetic structure, which is an antiferromagnetic structure suggested by Allen [6, 7].

Table 1 compares the preliminary results of our investigation. The oxygen distortion does not seem to reduce the total energy in either case (the 3k and 1k). The 3k and 1k structures appear to be almost degenerate in energy.

Structure	Energy Difference (mRy)
1k + SOC	0.00
3k transv. 2 + SOC	0.587
3k kmg + SOC	0.640
3k transv. 1 + SOC	0.655
1k + Dist + SOC	1.929
3k transv. 2 + Dist+SOC	3.388
3k transv. 2 + Dist+SOC	3.405

Table 1.
Energy differences with respect to the energy of the 1k magnetic structure with spin-orbital coupling (SOC).

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Fig. 2.
The figure displays the 3k magnetic structure (longitudinal). Only the U ions are shown for clarity. The moments point along the (111) direction.